The Orbital Selective Mott Transition in a Three Band Hubbard model: a Slave Boson Mean Field Study

Xi Dai[a,b], Gabriel Kotliar[c], Zhong Fang[a]

[a] Institute of Physics, Chinese Academy of Sciences, Beijing, China

[b]Department of Physics, University of Hong Kong,Hong Kong, China[*] and

[c]Department of Physics and Center for Material Theory,

Rutgers University, Piscataway, NJ 08854, USA

(Dated: February 6, 2008)

Abstract

In the present paper, we systematically studied the possible Orbital selective Mott transition (OSMT) in the t_{2g} system, which has three orbital degeneracy. The slave Boson mean field theory is generalized to the three band systems with full Hund's rule coupling including spin flip and pair hopping terms. A new type of Mott transition which is driven by the crystal field splitting or the lattice distortion is found is this model. We argue that this new type of Mott transition may relate to the puzzling phase in $Sr_xCa_{2-x}RuO_4$.

---

[*]E-mail:`daix@hkucc.hku.hk`

I. INTRODUCTION

The charge, spin, orbital and lattice are the main ingredient of the physics in transition metal oxides[1]. The interplay and competition among them are the key points to understand the interesting physical properties in transition metal oxides. On the other hand, Mott transition plays an important role in understanding the correlation effect in the solid state[2]. One of the important debating issues in the study of Mott transition is wether the Mott transition in realistic material is purely interaction driven or the orbital and lattice degrees of freedom also play some role[3, 19]. Although most of the theoretical studies on the Mott transition are based on the single band Hubbard model, most of the realistic materials which undergo Mott transition have more than one active orbit[2, 5]. The recent studies show that the Mott transition in the multi-orbital system is not the simple generation of its single band version, instead it has some interesting features which are unique in the multi-orbital case[4, 6]. It is even more complicated if the lattice distortion get involved and play some role in the Mott transition. Therefore it is very interesting to study how the orbital fluctuation and lattice distortion will affect the nature of Mott transition. Is there any new physics near the Mott transition point induced by the interplay between the charge, spin, orbital and lattice?

The orbital selective Mott transition (OSMT) is a very important feature for the Mott transition in the multi-orbital system and is first proposed by Anisimov in the study of $Sr_xCa_{2-x}RuO_4$.[5, 7–9] The orbital degree of freedom is involved in the OSMT and the Mott transition in different orbital happens individually because of the lift of the orbital degeneracy. In crystal, basically there are two origins which can induce the asymmetry in the orbital space. The first one is the breaking of the local rotational symmetry in the crystal, which makes the different orbits pointing along different direction have different band width. The second one is the crystal field splitting generated by the lattice distortion, which removes the degeneracy of the energy levels. A two-band generalized Hubbard model with half filling has been proposed[11, 12] to study the basic features of OSMT. Most of the studies on this two-band model using the dynamical mean field theory (DMFT)[13] reach the following common conclusions[11, 12, 14]. i)The OSMT is induced by the band width asymmetry of the different orbits. The crystal field splitting will reduce rather than enhance the tendency to OSMT. ii)The orbital selective Mott Phase (OSMP), in which one orbit is

already localized while the other one keeps itinerary, will be greatly enhanced when the symmetry of the local interaction is lowered by including the Hund's rule coupling terms.

Although the two-band model has been extensively studied in recent years, it may not be directly applied to the situation of $Sr_xCa_{2-x}RuO_4$, because the effective model here is the three-band model with four total electrons[5, 10]. The most interesting feature of the OSMT in $Sr_xCa_{2-x}RuO_4$ is that the OSMT will be accompanied by the change of orbital polarization. Before the OSMT, as predicted by LDA calculation and conformed by ARPES [9, 10], the four electrons are almost evenly distributed among the three t_{2g} orbits. Therefore the average occupation number of each orbital is 4/3 before the OSMT. In order to have OSMP, 1/3 of the electron has to be moved from the localized orbital to the non-localized ones. Thus the OSMT is always accompanied by the charge redistribution or the change of orbital polarization. Since the crystal field splitting strongly couples to the orbital polarization, it will play a very important role in the OSMT for the three-band model.

From the methodology point of view, it is very difficult to apply the DMFT to the three band Hubbard model, because of the limitation of the computation ability. In the present paper, we generalize the slave Boson mean field theory[15, 16] which is very successful in the single band Hubbard model to the three band model with both local repulsive interaction and Hund's rule coupling. The slave boson method is closely related to the Gutzwiller approximation, which was recently extended to incorporate the effects of multiplets[17]. We give the detail description of the slave Boson mean field theory in section II and benchmark it in the two band model with other approaches in section III. Finally we discuss the complete phase diagram for the three band Hubbard model with Hund's rule coupling in section IV and make some important conclusions in section V.

II. THE HAMILTONIAN AND THE SLAVE BOSON MEAN FIELD APPROXIMATION

The general Hamiltonian describing the t_{2g} system can be written as

$$
\begin{aligned}
H_{Total} &= H_0 + H_I \\
H_0 &= \sum_{k,\alpha\sigma} \epsilon_{k,\alpha} C^\dagger_{k\alpha\sigma} C_{k\alpha\sigma} + \sum_{k,\alpha\sigma} \Delta_\alpha C^\dagger_{k\alpha\sigma} C_{k\alpha\sigma} \\
H_I &= U \sum_{i,\alpha} n_{i\alpha\uparrow} n_{i\alpha\downarrow} + U' \sum_{i,\sigma\sigma',\alpha<\beta} n_{i\alpha\sigma} n_{i\beta\sigma'} - J_z \sum_{i,\alpha<\beta,\sigma} n_{i\alpha\sigma} n_{i\beta\sigma} \\
&\quad - J_{xy} \sum_{i,\alpha<\beta} \left[C^\dagger_{i\alpha\uparrow} C_{i\alpha\downarrow} C^\dagger_{i\beta\downarrow} C_{i\beta\uparrow} + C^\dagger_{i\alpha\downarrow} C_{i\alpha\uparrow} C^\dagger_{i\beta\uparrow} C_{i\beta\downarrow} \right] \\
&\quad - J_{xy} \sum_{i,\alpha<\beta} \left[C^\dagger_{i\alpha\uparrow} C^\dagger_{i\alpha\downarrow} C_{i\beta\uparrow} C_{i\beta\downarrow} + C^\dagger_{i\beta\uparrow} C^\dagger_{i\beta\downarrow} C_{i\alpha\uparrow} C_{i\alpha\downarrow} \right]
\end{aligned}
\tag{1}
$$

where α and σ are the orbital and spin indices respectively. U describes the on-site Coulomb interaction term between two electrons in the same orbit but with opposite spin. While U' describes the on-site interaction term for two electrons reside in different orbits. Δ_α represent the crystal field splitting of the three t_{2g} orbits. We divide the Hund's rule coupling terms into three parts. The first one is the J_z term which describes the longitudinal part of the Hund's rule coupling which only involves density-density coupling. While the other two J_{xy} terms describe the spin flip and pair hopping processes respectively. In the present study, we assume the system has approximately the cubic symmetry which gives the following constraint $U = U' + 2J$ with $J_z = J_{xy} = J$.

Following reference [15] and [16], we first diagonalize the local Hamiltonian H_I by a set of local bases $|\mu\rangle$ and for each of them we define a slave boson operator l^+_μ. Then we can express the physical electron operator in terms of the pseudo fermions $f^+_{i\alpha\sigma}$ and slave bosons as

$$
C^+_{i\alpha\sigma} = Z^+_{i\alpha} f^+_{i\alpha\sigma}
\tag{2}
$$

,

where

$$
Z^+_{i\alpha\sigma} = \sum_{\mu\nu} D^{\alpha\sigma*}_{\nu\mu} L_{i\alpha\sigma} l^+_{i\mu} l_{i\nu} R_{i\alpha\sigma}
\tag{3}
$$

with

$$L_{\alpha\sigma} = \frac{1}{\sqrt{n_{i\alpha\sigma}}} \tag{4}$$

$$R_{\alpha\sigma} = \frac{1}{\sqrt{1 - n_{i\alpha\sigma}}}$$

and $D_{\nu\mu}^{\alpha\sigma} = \langle \nu | C_{\alpha\sigma} | \mu \rangle$ being the matrix elements of the electron operator represented in terms of the local atomic states.

Therefore the original Hamiltonian can be written in terms of the slave particles as

$$H_{sb} = \sum_{ij,\alpha\sigma} t_{ij,\alpha\sigma} f_{i\alpha\sigma}^+ f_{j\alpha\sigma} Z_{i\alpha\sigma}^+ Z_{j\alpha\sigma} + H.C. + \sum_{i,\alpha\sigma} \Delta_\alpha f_{i\alpha\sigma}^+ f_{i\alpha\sigma} + \sum_{i,\mu} E_\mu l_{i\mu}^+ l_{i\mu} \tag{5}$$

with two following local constraints

$$\sum_\mu l_{i\mu}^+ l_{i\mu} = 1 \tag{6}$$

$$\sum_\mu \eta_{\alpha\sigma,\mu} l_{i\mu}^+ l_{i\mu} = f_{i\alpha\sigma}^+ f_{i\alpha\sigma} \tag{7}$$

, where $\eta_{\alpha\sigma,\mu} = \sum_\nu D_{\nu\mu}^{\alpha\sigma *} D_{\nu\mu}^{\alpha\sigma}$ is the beverage particle number with the orbital α and spin σ for the configuration μ.

In the mean field approach, we treat all the boson operators l_μ and l_μ^+ as c-numbers which will be determined by minimization the ground state energy. The local constraints 6 and 7 will be released to be global ones, which can be satisfied by two Lagrange multiples λ_1 and λ_2. Therefore we obtain the following mean field Hamiltonian

$$H_{mf} = \sum_{ij,\alpha\sigma} t_{ij,\alpha\sigma} f_{i\alpha\sigma}^+ f_{j\alpha\sigma} Z_{\alpha\sigma}^* Z_{\alpha\sigma} + H.C. + \sum_{i,\alpha\sigma} (\Delta_\alpha - \mu_f + \lambda_1) f_{i\alpha\sigma}^+ f_{i\alpha\sigma} \tag{8}$$

$$+ \sum_{i,\mu} (E_\mu - \xi_\mu) l_\mu^+ l_\mu + \sum_{i,\mu} \lambda_2 \left(l_\mu^+ l_\mu - 1 \right)$$

with $\xi_\mu = \sum_{\alpha\sigma} \eta_{\alpha\sigma,\mu} \lambda_1$. The mean field solution can be determined by the stationary condition of the ground state energy,

$$\frac{\partial E \left[l_\mu, l_\mu^+, \lambda_1, \lambda_2 \right]}{\partial l_\mu} = 0$$

$$\frac{\partial E \left[l_\mu, l_\mu^+, \lambda_1, \lambda_2 \right]}{\partial \lambda_i} = 0 \tag{9}$$

For the three band model discussed here, we have to minimize the mean field ground state energy respect to all the 64 independent l_μ as well as two Lagrange multipliers and one chemical potential. Therefore the main difficulty here is how to reach the saddle point efficiently in the 67 dimensional parameter space. The simple iterative procedure is very hard to converge and is almost useless in practice. In the present paper, we solve this problem by a so called "adiabatic solution searching" procedure. First we solve the mean field self consistent equation for a symmetric 3-band model with equal band width, no crystal field splitting and $J = 0$ ($SU(6)$ symmetric case), which is easy due to the high degeneracies of the local configurations. Then we tune the parameters step by step towards the realistic parameters we are studying. For each step, we use the converged solution of the previous step as the initial value for the iteration. Since we only change the parameters slightly at each step, the new self consistent solution should be very close to the previous one and the convergence can be reached very soon. Therefore when the parameters evolute slowly to their actual values, we adiabatically obtained the solution.

III. BENCHMARKS

In this section, we benchmark our present mean field approach with other numerical approaches. First we apply the slave Boson mean field theory to the multi-band Hubbard model with SU(N) symmetry, where both the crystal field splitting and Hund's rule coupling has been set to zero. The non-interacting density of states of each band is chosen to be semi-circle with half bandwidth D, which also applies to all the models studied in this paper. The quasi-particle weight reduction as the function of U/D is plotted in figure 1. The Mott transitions can be seen clearly where the quasi-particle weight vanishes. The critical interaction U_c for 1,2 and 3 band model are found to be $3.42D$, $5.1D$ and $6.27D$, which is in good agreement with the results obtained by dynamical mean field theory (DMFT) and other slave particle mean field theories[14].

We then apply the slave boson mean field approach to the 2-band Hubbard model with unequal bandwidth D_2 and D_1 without crystal field splitting. The total occupation number of the electrons are fixed to be two, which keeps the particle hole symmetry for this model. The DMFT study on this model reveals an interesting new phase called orbital selective Mott phase (OSMP), where the narrow band is in the Mott insulator phase while the wide
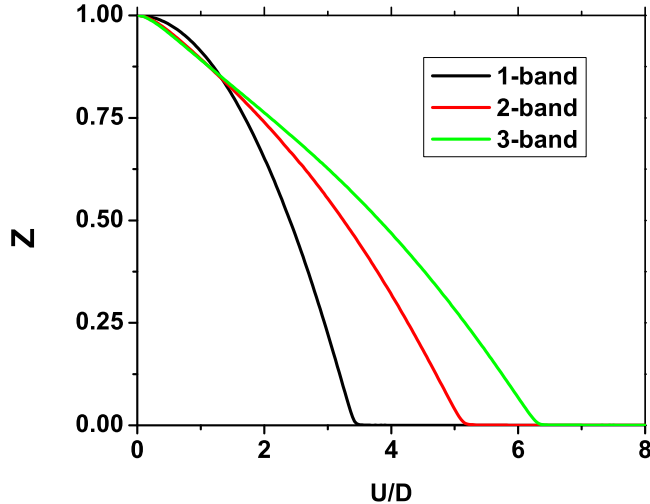
FIG. 1: The quasi-particle weight as the function of U/D for 1,2, and 3 band Hubbard model.

band still remains metallic[11, 12]. This simplified model is first proposed to explain the possible orbital selective Mott transition in the $Ca_{2-x}Sr_xRuO_4$. Since it has been widely studied by DMFT as well as mean field approaches, it is quite suitable to be the benchmark of our approach. The DMFT studies on this model show that the OSMT does not occur without the Hund's rule coupling term for $D_1/D_2 = 0.5$. This is because the local interaction has the $SU(4)$ symmetry without the Hund's rule coupling terms, which prevents the OSMT from occurring due to enhancement of the orbital fluctuation. We plot the phase diagram obtained by the slave boson mean field theory in figure 2 with y-axis being the interaction strength and x-axis being the radio of two band width.

We found that the OSMP appears only when the bandwidth ratio is below a certain critical value, which is found to be around $D_2/D_1 < 0.25$. This result is in good agreement with that of the slave spin mean field theory[14]. The role of the Hund's rule coupling has also been discussed by many groups within the frame of DMFT[11, 12]. With the increment of the Hund's rule coupling, the area of the OSMP in the phase diagram get larger and larger. In Figure 3 we plot the phase diagram with the full Hund's rule coupling including both the spin flip and pair hopping terms in equation 1 .

The transverse part of the Hund's rule coupling including the spin-flip and pairing hop-
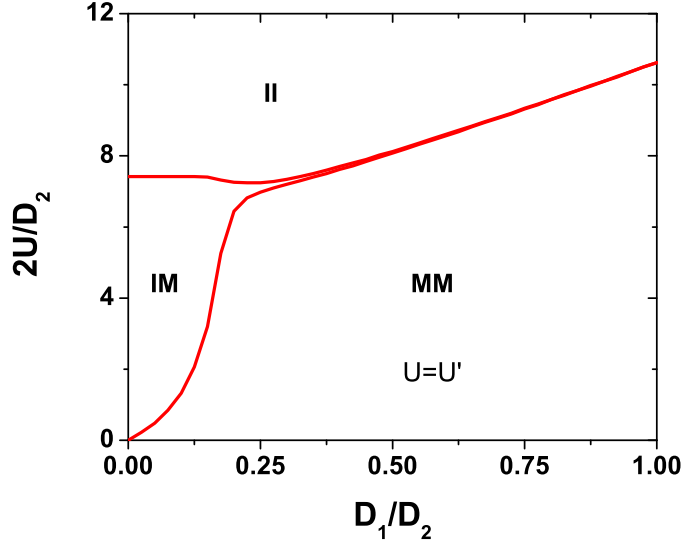
FIG. 2: The phase diagram for the 2-band Hubbard model without Hund's rule coupling.

ping terms plays an extremely important role in OSMT. The OSMP is strongly suppressed without these two transverse terms. This important feature can be also captured by the present slave boson mean field theory. As shown in Figure 4, the area of OSMP is shrunk dramatically when the two transverse terms are switched off, which is also in good agreement with the DMFT results.

IV.   OSMT IN THE THREE BAND HUBBARD MODEL

The theoretical study of the orbital selective Mott transition is motivated by the surprising Curie-Weiss behavior of the spin susceptibility in CaSrRuO4, which is unexpected because the transport measurement shows metallic behavior. The two band Hubbard model with Hund's rule coupling is proposed as the simplest toy model for OSMT. While compared to the realistic situation in CaSrRuO4, which has four electrons occupying three t2g orbitals, the two band model is over simplified and can not capture a very important feature of OSMT in $Sr_xCa_{2-x}RuO_4$. Unlike the situation in the 2 band model, the OSMT in $Sr_xCa_{2-x}RuO_4$ is accompanied by the charge redistribution even without crystal field splitting. Since the average occupation number in $Sr_xCa_{2-x}RuO_4$ is 4/3 per orbital per site, which is away from
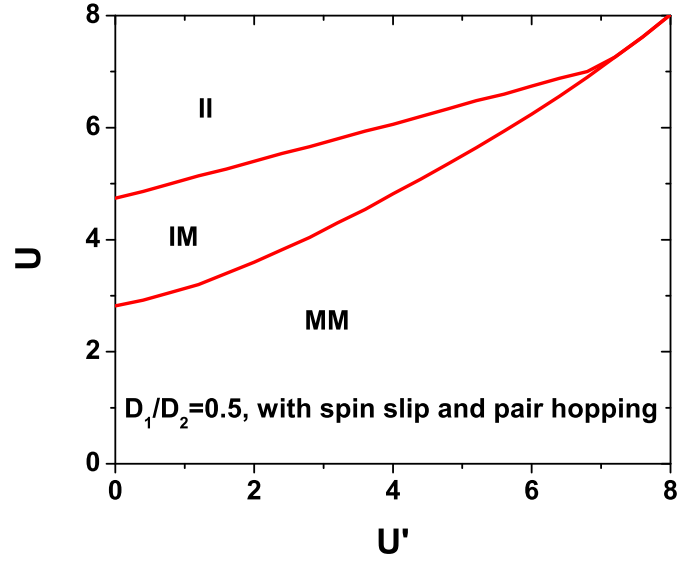
8

FIG. 3: The phase diagram for the 2-band Hubbard model with the full Hund's rule coupling.

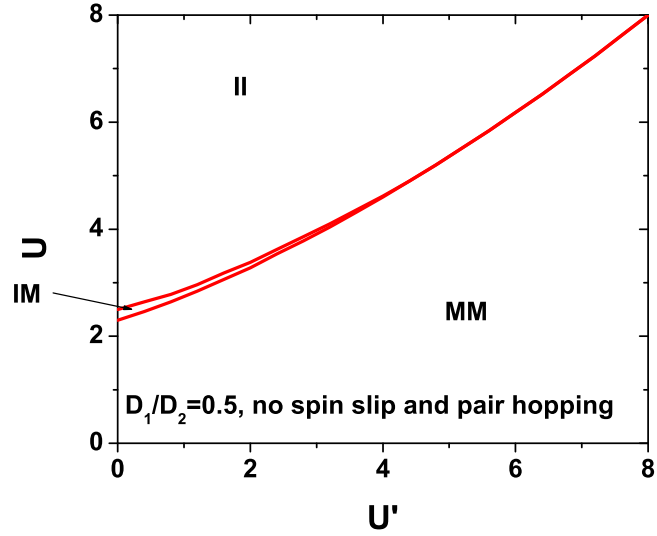
FIG. 4: The phase diagram for the 2-band Hubbard model with only longitudinal terms in Hund's rule coupling.

the half filling, 1/3 of electron has to be moved from the wide bands to the narrow band to make it half filled and insulating. Therefore the crystal field splitting, which always reduces the tendency of OSMT in the two band model, can greatly enhance the OSMT in the three band model, because it can induce such charge redistribution required by OSMT. In the present paper, we have systematically studied the three-band Hubbard model with Hund's rule coupling using the slave boson mean field approach. Since the optical conductivity measurement[7] indicate the local moment is on the d_{xy} orbital, we choose the band width of the three different bands in eq.1 to be $2D, 2D$ and D. The phase diagram for the full Hund's rule coupling is plotted in figure 5 with the interaction strength U and crystal field splitting $E = \Delta_1 - \Delta_3$ being the y and x axis respectively. The $(3,1)II$ phase represents the Mott insulator phase for both wide and narrow bands with occupation numbers of wide band and narrow band to be 3 and 1 respectively. $(3,1)MI$ phase represent the metallic phase for the wide band and insulating phase for the narrow band. MM phase represents the metallic phase for all the three bands. And $(4,0)II$ phase represents the band insulator phase with all the four electrons filling the two wide bands. We find that the OSMT never occurs for this model without crystal field splitting. Small crystal field splitting will induce an OSMT very effectively indicating that the metallic state is unstable against the crystal field in this regime. Since the crystal field splitting is induced by lattice distortion, our result indicates that the OSMT in $Sr_xCa_{2-x}RuO_4$ is not purely interaction driven. Instead, the lattice degree of freedom will also paly a very significant role. To further verify this point, we plot the quasi-particle weight as the function of the amount of charge transfer among t_{2g} orbits, which is determined by the crystal field splitting, in figure7. We can see very clearly that the quasi-particle weight goes to zero when the charge transfer δ reaches 1/3, which indicates a new type of Mott transition driven by the crystal field splitting. When switching off the spin flip and pair hopping terms, the over all feature of the phase diagram keeps unchanged, especially the OSMT does not occur without the crystal field splitting. The area of OSMP in the phase diagram decreases dramatically as shown in figure 6 which is quite similar to the two band model.
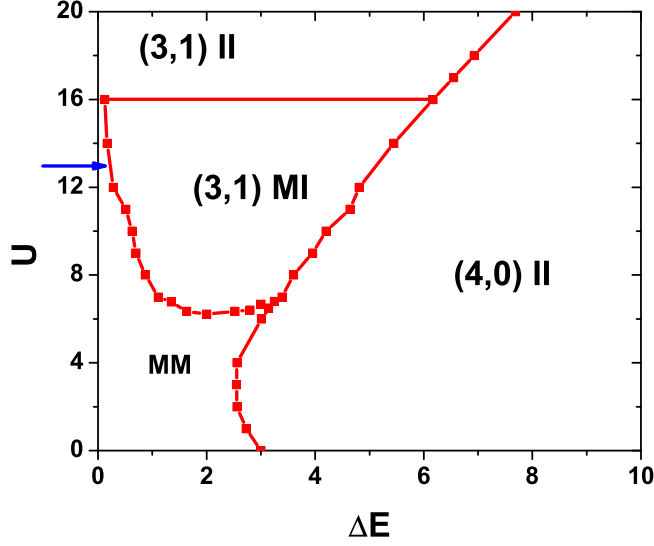
FIG. 5: The phase diagram for the OSMT in the 3-band Hubbard model with full Hund's rule coupling $J = 0.25U$. The x-axis is the crystal field splitting and y-axis is the intra band local interaction U.

V.   CONCLUDING REMARKS

The OSMT in three-band Hubbard model with different band width is studies in detail using the slave Boson mean field theory. First we generalize the slave Boson approach proposed by G. Kotliar and A. E. Ruchenstein[15] to the multi-band system with full Hund's rule coupling terms and benchmark it with the other numerical approaches like DMFT in the two band Hubbard model. Then we apply it to study the possible OSMT in the three-band Hubbard model with both Hund's rule coupling terms and crystal field splitting. Our numerical result shows that unlike the case in the two-band model the crystal field splitting plays a very important role in stabilizing the OSMP. In fact a new type of OSMT, which is driven by the crystal field is observed in our study. Since the crystal field splitting is induced by the lattice distortion, our results strongly suggest that the lattice degree of freedom is plays a crucial role in the OSMT in the compound like $Sr_xCa_{2-x}RuO_4$.
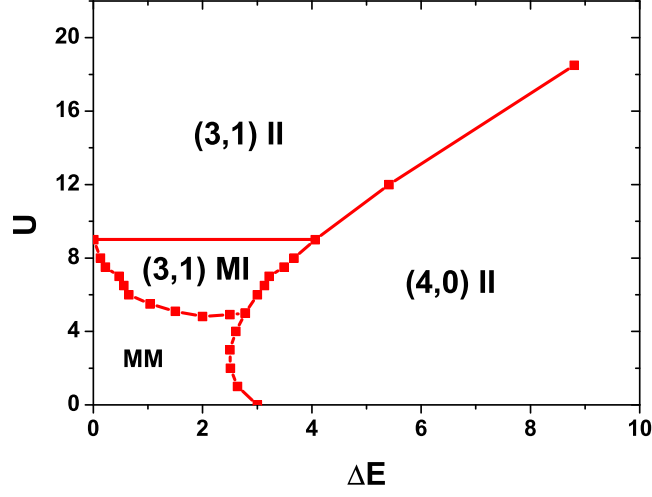
11

FIG. 6: The phase diagram for the OSMT in the 3-band Hubbard model with only longitudinal Hund's rule coupling $J = 0.25U$. The x-axis is the crystal field splitting and y-axis is the intra band local interaction U.
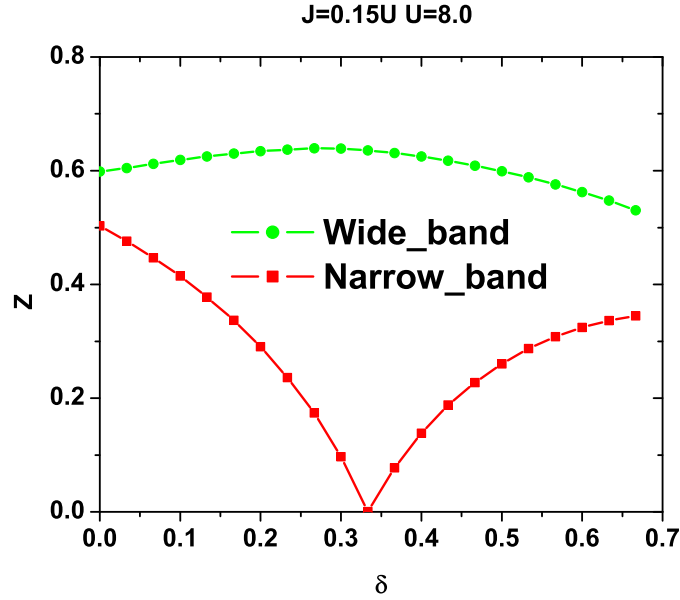

FIG. 7: The quasi-particle weight for the wide band (circles) and narrow band (squares) as the function of δ, where $\delta = n_{narrow} - \frac{4}{3}$.

[1] Y. Tokura and N. Nagaosa, SCIENCE **288**, 462, (2000).

[2] M. Imada, A. Fujimori, and Y. Tokura, Rev. Mod. Phys. **70**, 1039 (1998).

[3] S. Biermann, A. Poteryaev, A. I. Lichtenstein, and A. Georges Phys. Rev. Lett. **94**, 026404 (2005).

[4] M. Capone, M. Fabrizio, C. Castellani and E. Tosatti, SCIENCE **296**, 2364, (2002).

[5] V. Anisimov, I. Nekrasov, D. Kondakov, T. Rice and M. Sigrist, Eur. Phys. J. B **25**, 191, (2002).

[6] G. Kotliar and H. Kajueter, Phys. Rev. B **54**, R14221 (1996); M. J. Rozenberg, Phys. Rev. B **55**, R4855 (1997); K. Held and D. Vollhardt, Eur. Phys. J. B **5**, 473 (1998); J. E. Han, M. Jarrell, and D. L. Cox, Phys. Rev. B **58**, R4199 (1998).

[7] S. Nakatsuji and Y. Maeno, Phys. Rev. Lett. **84**, 2666 (2000).

[8] J. S. Lee, S. J. Moon, T.W. Noh, S. Nakatsuji, and Y. Maeno, Phys. Rev. Lett. **96**, 057401 (2006).

[9] S.-C. Wang et al., Phys. Rev. Lett. **93**, 177007 (2004).

[10] Z. Fang, N. Nagaosa, and K. Terakura, Phys. Rev. B **69**, 045116, (2004).

[11] A. Koga, N. Kawakami, T.M. Rice, and M. Sigrist, Phys. Rev. Lett. **92**, 216402 (2004); A. Koga, K. Inaba, and N. Kawakami, cond-mat/0510449 (2005).

[12] A. Liebsch, Phys. Rev. Lett. **91**, 226401 (2003); A. Liebsch, Europhys. Lett. **63**, 97 (2003); A. Liebsch, Phys. Rev. B **70**, 165103 (2004).

[13] A. Georges, G. Kotliar, W. Krauth, and M. J. Rozenberg, Rev. Mod. Phys. **68**, 13 (1996).

[14] L. de'Medici, A. Georges, and S. Biermann, Phys. Rev. B **72**, 205124 (2005).

[15] G. Kotliar and A. E. Ruchenstein, Phys. Rev. Lett. **57**, 1362 (1986).

[16] R. Fresard and G. Kotliar, Phys. Rev. B **56**, 12909 (1997); H. Hasegawa, Phys. Rev. B **56**, 1196, (1997).

[17] J. Bunemann, W. Weber and F. Gebhard, Phys. Rev. B **57**, 6896 (1998);

[18] S. Florens, A. Georges, G. Kotliar, and O. Parcollet, Phys. Rev. B **66**, 205102 (2002).

[19] S. Okamoto and A. J. Millis, Phys. Rev. B **70**, 195120 (2004).